

**INVESTIGATION OF DEEP-WATER CIRCULATION MODES IN
THE EARLY CENOZOIC USING NEODYMIUM ISOTOPES FROM
FOSSIL FISH DEBRIS**

A Senior Scholars Thesis

by

LANDON BLAKE JONES

Submitted to the Office of Undergraduate Research
Texas A&M University
in partial fulfillment of the requirements for the designation as

UNDERGRADUATE RESEARCH SCHOLAR

April 2011

Major: Geology

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ABSTRACT

Investigation of Deep-water Circulation Modes in the Early Cenozoic using Neodymium Isotopes from Fossil Fish Debris. (April 2011)

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The ocean's deep-water circulation plays a large role in heat transport across the globe. Circulation in the modern begins where cold, dense surface waters of the North Atlantic and Southern oceans sink to form Atlantic Bottom water. However, this mode did not operate in the geologic past. A growing body of Nd isotope data from fossil fish debris is being used to reconstruct the ancient mode of deep-water circulation throughout the early Cenozoic greenhouse interval. Recent data from previous Ocean Drilling Program (ODP) sites suggest that a bipolar mode of meridional overturning circulation may have existed in the Pacific during the early Cenozoic, beginning ~65 million years ago and lasting until ~40 million years ago. Here I present new data from Deep Sea Drilling Project (DSDP) Site 464, Northern Hess Rise, to enhance the reconstruction of deep water mass composition as well as determine if a reductive cleaning step ("clean") method is necessary during sample preparation. Site 464 $\epsilon_{\text{Nd}}(t)$ values range from -.30 to less radiogenic values of -4.42 from ~56.0 to 32.3 million years ago, showing a shift from a North Pacific deep-water influence to a Southern Ocean influence. The

comparison of “clean” versus “unclean” analyses indicates that both record the same seawater composition.

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NOMENCLATURE

$^{\circ}\text{C}$	Degrees Celsius
Ma	Million years ago
Myr	Million years
ϵ_{Nd}	Nd isotopic composition of seawater
Nd	Neodymium

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CHAPTER I

INTRODUCTION

The oceanic thermohaline circulation plays a large role in the overall net heat transfer from equator to poles [e.g., *Broecker*, 1997], which in turn is a significant component of the overall global climate system. Throughout earth's history there have been different climate intervals that behaved in different manners; an important question is what role ocean circulation played in those different climates systems. In order to answer this we must understand how the thermohaline circulation of ancient seas operated.

In the modern oceans cold, dense surface waters from the North Atlantic and Southern Oceans sink to form deep waters and these waters circulate throughout the deep ocean basins ultimately returning to the surface in the North Pacific – the “Great Ocean Conveyor Belt” coined by [*Broecker*, 1987]. However, the mode of circulation likely was different during the last major “greenhouse” climate interval due to the vastly different climatic and tectonic boundary conditions during that time. Such different boundary conditions complicate our understanding of deep-water circulation patterns.

Over the last 65 Ma, earth's climate has experienced large scale transitions of climate extremes. There have been times of extreme “greenhouse” warmth with ice-free poles, to extreme “icehouse” conditions with massive polar ice caps [e.g. *Zachos et al.*,

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2001]. During the last climatic interval of greenhouse warmth, ~65 to 45 million years ago, the thermal structure of the oceans was fundamentally different from that of the modern oceans. High-latitude sea surface temperatures during the late Paleocene ~55 Ma were 9° to 11°C higher than present day SST and increased by 4° over the next 3 to 4 Myr; giving SST during the early Eocene to be 14° to 16°C. Deep-water temperatures of this interval were normally between 7° and 10° and increased during the early Eocene to between 10° and 14° [e.g. *Zachos et al.*, 1994]. Also, the equator to pole thermal gradient during the early Cenozoic was nearly half that of the Holocene [e.g. *Stott et al.*, 1990].

Equally important to the changing mode of deep-water circulation is the different tectonic boundary conditions that existed in the early Cenozoic. The paleogeography and ocean basin configuration that existed ~55 Ma for example, was significantly different than in the present. The Norwegian-Greenland and Labrador seas had just begin forming in the northern North Atlantic [e.g., *Talwani and Eldholm*, 1977; *Saunders et al.*, 1987] and the Tasman Sea and Drake Passage were not open [e.g., *Barker and Burrell*, 1977; *Lawver and Gahagan*, 2003]. It is unlikely that deepwater would have formed in the North Atlantic because the Greenland and Norwegian seas had not fully formed [e.g., *Saunders et al.*, 1997], causing waters in the Greenland-Scotland Ridge Region to be too warm to sink [e.g. *Zachos et al.*, 2001]. However, at lower latitudes there may have been exchange between the Pacific and Atlantic Ocean via the Caribbean because the Isthmus of Panama had not formed.

Data collected by [Tucholke and Miller, 1983; Mountain and Miller, 1992; Pak and Miller, 1992; Zachos *et al.*, 1992; Thomas *et al.*, 2003; Via and Thomas, 2006] give evidence supporting the Southern Oceans as the main region of deepwater formation during the early Cenozoic. As in the modern climate system, the Pacific likely exerted a dominant control on ancient climate. During the late Cretaceous and early Paleogene the Pacific basin was proportionately larger than the modern basin. This is because plate tectonic processes are gradually diminishing the size of the basin due to subduction. A larger Pacific likely would have impacted global climate more than the Atlantic or other basins.

Data collected by [Thomas, 2004] and [Thomas *et al.*, 2008] suggest that a bimodal production of deep waters existed from ~65 to 40 million years ago (Ma) that sourced from the high latitudes of the Northern and Southern hemispheres. Data collected from DSDP and Ocean Drilling Program (ODP) sites also indicate that the Atlantic waters had minimal if any effect on tropical Pacific deepwater [Thomas *et al.*, 2008]. However, these hypothesized models for deep-ocean circulation ~65-40 Ma are based on the data available at the time which provides relatively sparse geographic and bathymetric coverage.

Here I present new Nd isotope data from Deep Sea Drilling Project (DSDP) Site 464, from Hess Rise in the North Pacific to contribute to the growing reconstruction of Pacific deep-water mass composition (Figure 1). The new data corroborate the findings of [Thomas, 2004 and Thomas *et al.*, 2008], supporting the hypothesis that deep waters formed in the North Pacific during part of the early Paleogene. Furthermore these data

will contribute to models of how deep-ocean circulation may have impacted heat transport during “greenhouse” climate states.

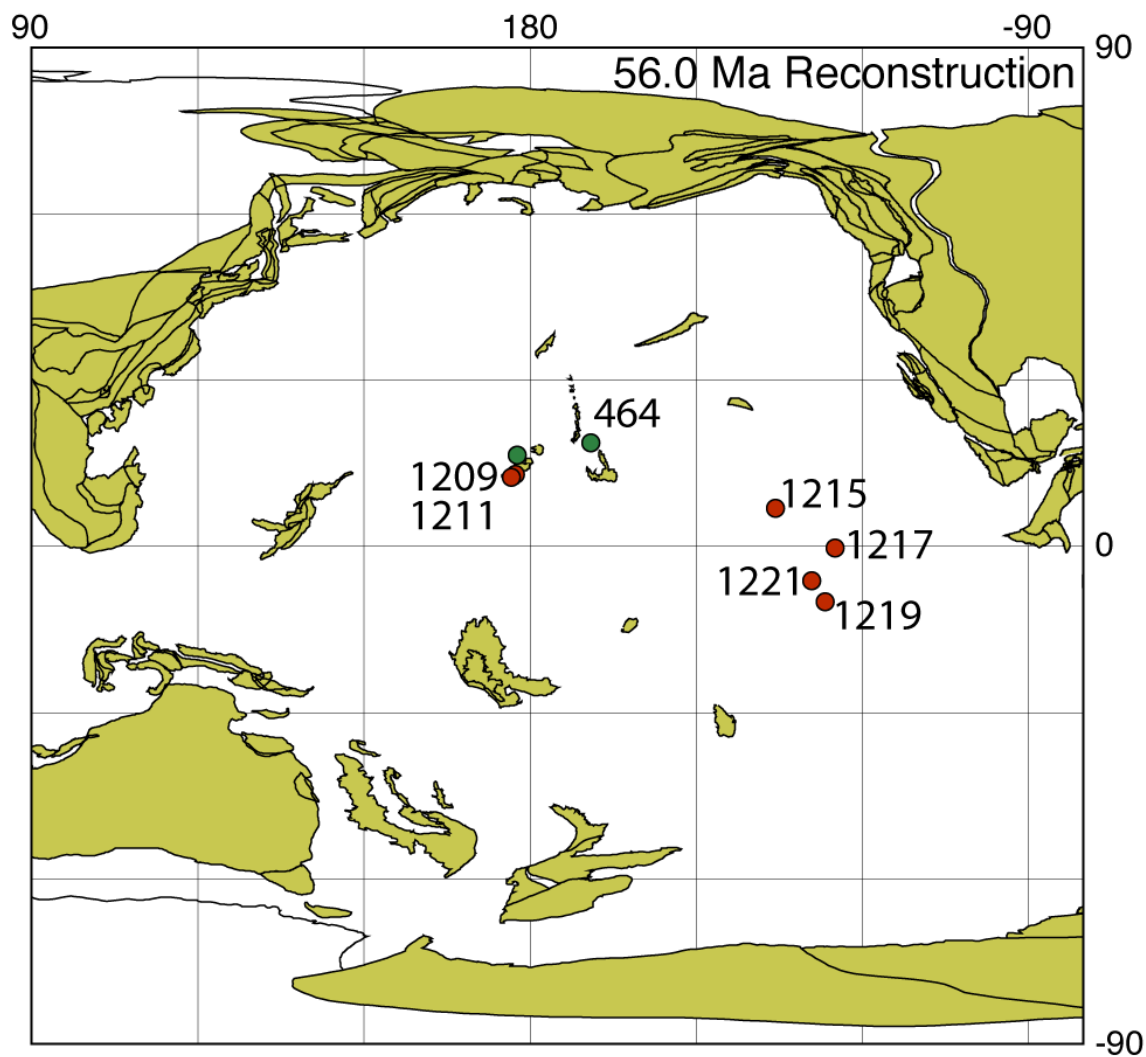


Figure 1. Paleogeographic reconstruction of 56 Ma (Ocean Drilling Stratigraphic Network, <http://www.odsn.de>) showing the location of ODP Leg 199 Sites and Site 464; which was investigated in this study.

CHAPTER II

METHODS

2.1. Nd Isotopes used as Paleoceanographic Indicators

Neodymium isotopic compositions in seawater are used as tracers for water mass composition to study ancient deep-water circulations patterns. Neodymium is a rare earth element that has seven naturally occurring isotopes. This study employs the ratio of the radiogenic isotope ^{143}Nd , produced by the decay of the ^{147}Sm , to the stable isotope ^{144}Nd . The half-life of ^{147}Sm is 1.06×10^{11} years, thus variations of $^{143}\text{Nd}/^{144}\text{Nd}$ values are small. The ratio of $^{143}\text{Nd}/^{144}\text{Nd}$ in a given lithology is a result of varying Sm and Nd concentrations obtained during the initial formation of minerals. Nd isotope data are presented in epsilon notation which is the $^{143}\text{Nd}/^{144}\text{Nd}$ value of the sample normalized to the bulk earth composition, CHUR, where:

$$\epsilon_{\text{Nd}} = ((^{143}\text{Nd}/^{144}\text{Nd}_{\text{sample}}/0.512638) \times 10,000) - 1$$

[e.g., *DePaolo and Wasserburg, 1976*]. Low $^{143}\text{Nd}/^{144}\text{Nd}$ values convert to negative, nonradiogenic ϵ_{Nd} values; these are found in old, continental rocks. Higher $^{143}\text{Nd}/^{144}\text{Nd}$ result in higher ϵ_{Nd} values; these are found in younger, mantle derived rocks such as mid-ocean ridge basalts [e.g., *DePaolo and Wasserburg, 1976*].

The supply of Neodymium to the oceans is a result of weathering and drainage of subaerially exposed rocks, which leads to different variations of seawater ϵ_{Nd} [*Goldstein and Jacobsen, 1987*]. The isotopic composition of the surface waters sink in areas of water mass subduction and downwelling which impart the isotopic composition to intermediate and deepwater masses [*Goldstein and Jacobsen, 1987; Elderfield et al.,*

1990; *Halliday et al.*, 1992; *Sholkovitz*, 1993]. During subsequent circulation, the initial ϵ_{Nd} of a given water mass can be altered through mixing with other water masses [e.g., *Piepgras and Wasserburg*, 1982, 1987; *Piepgras and Jacobsen*, 1988; *Jeandel*, 1993; *Shimizu et al.*, 1994; *Jeandel et al.* 1998; *Amakawa et al.*, 2000]. Changes in weathering inputs from the continents along with different sources of deep waters can cause temporal variations in the deep-water Nd composition at a location.

Weathering and drainage of ancient Canadian rocks give the most negative, nonradiogenic ϵ_{Nd} values in the North Atlantic to be approximately -12 to -14, while weathering and drainage of the Pacific arc terranes give the most radiogenic values in the North Pacific to be approximately -5. The Indian and Southern Oceans are composed of ϵ_{Nd} values that lie between the end members of the North Atlantic and North Pacific [*Jones et al.*, 1994]. The interbasinal differences discussed here are a result of the oceanic residence times of Nd (~ 1500 yr [*Broecker et al.*, 1960]) being short compared to overall ocean mixing times (~ 1000 yr [e.g., *Tachikawa et al.*, 1999]). The short oceanic residence time of Nd allows the element to be a useful tracer of deep-water circulation [e.g., *Piepgras and Wasserburg*, 1982; *Bertram and Elderfield*, 1993; *Jeandel*, 1993].

The dissolved materials that drain into source regions give the individual deep-water mass its Nd isotopic composition [*Goldstein and Jacobsen*, 1987; *Elderfield et al.*, 1990; *Sholkovitz*, 1993]. An example of this is North Atlantic Deep Water (NADW); this is dense water that forms in the Nordic Seas with a ϵ_{Nd} signature of ~ -12 . The water flows south, mixing with sinking waters of the Labrador Seas with a ϵ_{Nd} signature of $\sim -$

26 which results in a water mass with a ϵ_{Nd} signature of $\sim -12 - 13$. The deep-sea transit of North Atlantic Deep Water can then be traced from the resulting water mass [Piepgras and Wasserburg, 1987]. Other waters, such as Antarctic Intermediate Water and Antarctic Bottom Water tend to be more radiogenic than North Atlantic Deep Water. These waters, classified as Southern Ocean waters, are formed by the mixing of eastward flowing waters through the Drake Passage with North Atlantic Deep Water, which gives Antarctic water a ϵ_{Nd} signature of ~ -9 [Piepgras and Wasserburg, 1982].

In order to exploit the marine geochemical cycling of Nd to reconstruct ancient water mass composition, we need a phase that accumulates in marine sediments that records and retains the deep-water Nd isotopic composition. The teeth and bones of fossil fish present such a phase. These materials acquire high Nd concentrations, 100-1000ppm, at the seafloor [e.g., Wright *et al.*, 1984; Shaw and Wasserburg, 1985; Staudigel *et al.*, 1985]). However, it must be noted that living fish do not contain Nd therefore, where fish swim and what they eat do not affect the overall Nd isotopic composition found in their fossilized remains [e.g., Wright *et al.*, 1984; Staudigel *et al.*, 1985]. Instead, fish debris acquire their Nd signature during early diagenesis at the sediment/ water interface [e.g., Staudigel *et al.*, 1985] thus, the overlying bottom water's Nd composition is recorded from the fossilized material at the seafloor. Data collected by [Martin and Haley, 2000; Martin and Scher, 2004] and confirmed by [Thomas, 2005] show that the Nd isotopic composition of fish teeth is not affected by late stages of diagenesis and therefore maintains its primary, deep-water mass signal throughout burial.

2.2. Analytical details

The ten samples used for my project were taken from DSDP Site 464 which is presently stored at the Integrated Ocean Drilling Program's Gulf Coast Repository at Texas A&M University. I washed the samples over a 63 μ m sieve, retained the coarse (>63 μ m) fraction, and dried them in a 50 °C oven. I hand-picked fish debris from the >63 μ m fraction, dividing the debris from each sample into two splits: "clean" and "unclean". The "clean" fish debris was subjected to a rigorous oxidative/reductive cleaning process outlined by [Boyle, 1981] while the "unclean" debris was only rinsed twice with ethanol and twice with ultrapure water. This test is to confirm that both initial steps yield the same isotopic composition.

Once the initial cleaning steps were completed the samples were dissolved in 2N HNO₃, and run through two separate column separation procedures. The first step employed RE Spec columns which separate the bulk REE from the other elements. Then the second step involved methylactic acid column chemistry to separate the Nd from the other REEs found in the sample. The samples consisting of only Nd were then analyzed by the thermal ionization mass spectrometry using the Thermo Triton in the R. Ken Williams ⁴⁵ Radiogenic Isotope Geosciences Laboratory in the College of Geosciences.

CHAPTER III

RESULTS

Of the 18 original samples, 13 samples from DSDP Site 464 (Northern Hess Rise) ran successfully (Table 1, Figure 2). The data fall into two categories: 10 “uncleaned” samples that make up the time series, and 3 “cleaned” samples (3 replicates of “uncleaned” samples). Comparison between the “cleaned” and “uncleaned” analyses from samples 6-2, 97-99cm, 6-6, 64-66cm, and 7-1, 50-52cm indicates that each fraction yielded similar values (Table 1). The difference between the two analyses for 6-2, 97-99cm was 0.02 epsilon units, that for 6-6, 64-66cm was 0.53, and the difference was 0.16 for sample 7-1, 50-52cm. Only the middle sample had a “cleaned” vs. “uncleaned” difference greater than analytical error. The “uncleaned” $\epsilon_{Nd}(0)$ values vary between -0.76 and -4.76. These data generally decrease from the base of the study interval to the top of the section investigated; however there are some finer scale changes superimposed on this general trend.

The $\epsilon_{Nd}(0)$ values for the “unclean” samples increased from -1.63 at 53.50 meters below sea floor (mbsf) to -.76 at 53.10 mbsf and then decrease to -2.61 at 51.50 mbsf. From there the $\epsilon_{Nd}(0)$ values increase to -2.32 at 49.64 mbsf, then decrease to -4.76 at 45.50 mbsf which is the highest recorded epsilon value in the section. This is followed by another increase to -3.32 at 43.97 mbsf, followed by a decrease to -4.65 at the top of the measured section.

Table 1. Site 464 Nd isotope data generated for this study. (*) indicates “cleaned” sample.

Depth (mb.s.f.)	Age (Ma)	$\epsilon_{\text{Nd}}(0)$	Error	$^{143}\text{Nd}/^{144}\text{Nd}$	%Standard Err	Abs Err	$\epsilon_{\text{Nd}}(t)$
42.40	32.35	-4.65	0.04	0.51239957	0.00044	0.0000023	-4.38
43.97	36.43	-3.32	0.03	0.51246760	0.00033	0.0000017	-3.02
*43.97	36.43	-3.38	0.04	0.51246448	0.00047	0.0000023	-3.08
45.50	40.41	-4.76	0.04	0.51239400	0.00035	0.0000018	-4.42
46.80	43.78	-3.39	0.03	0.51246447	0.00029	0.0000015	-3.02
48.55	48.33	-3.29	0.08	0.51246959	0.00085	0.0000043	-2.88
49.64	51.17	-2.32	0.03	0.51251923	0.00031	0.0000016	-1.89
*49.64	51.17	-1.79	0.06	0.51254634	0.00060	0.0000030	-1.36
51.50	54.97	-2.61	0.03	0.51250440	0.00031	0.0000016	-2.15
*51.50	54.97	-2.45	0.04	0.51251247	0.00034	0.0000018	-1.99
52.25	55.37	-2.18	0.03	0.51252624	0.00031	0.0000016	-1.72
53.10	55.82	-0.76	0.10	0.51259885	0.00100	0.0000053	-0.30
53.50	56.04	-1.63	0.03	0.51255436	0.00034	0.0000017	-1.16

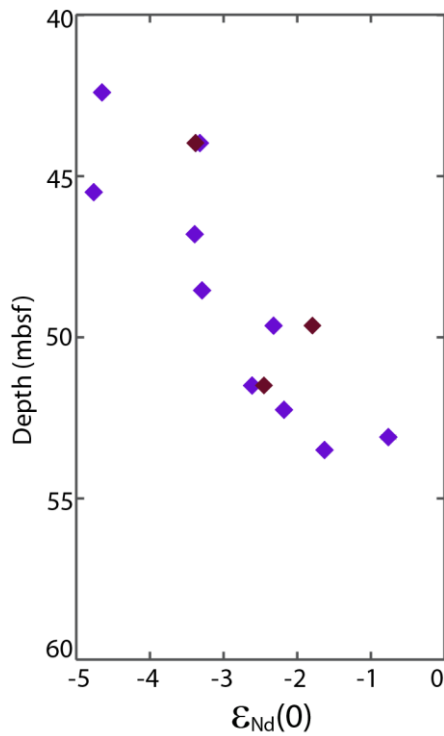


Figure 2. Nd isotope data generated from DSDP Site 464. Data are plotted verse meters below sea floor (mbsf). Clean samples are colored red.

CHAPTER IV

SUMMARY AND CONCLUSIONS

4.1. Comparison of cleaned and uncleaned samples

A growing body of work suggests that the accepted protocol for cleaning fish teeth and bones prior to dissolution and column chemistry – the rigorous oxidative/reductive cleaning process outlined by [Boyle, 1981] – may not be necessary. This is because the oxide coating that is removed during the cleaning process records the same isotopic signal as the biogenic apatite. However the studies that suggest the cleaning is unnecessary investigated relatively recent intervals of time. In order to demonstrate that the cleaning was not needed for more ancient samples, I performed a comparison of three sample pairs (a “cleaned” and an “uncleaned” fraction from three different samples). Comparison of samples subjected to both methods indicates that cleaned and uncleaned samples yield the same Nd isotope ratios (Figure 2). While the difference between the cleaned and uncleaned splits for sample 6-6, 64-66cm was ~0.5 epsilon units (and greater than the external precision), this difference still falls within the range of natural intra-sample variability demonstrated for populations of fish debris within a given 2 cm sampling interval [Thomas and Via, 2007]. Thus, elimination of the time consuming and hazardous cleaning step is justified even for samples older than 50 million years. The data collected from uncleaned samples may therefore be used for interpretation of deep-water mass circulation.

4.2. Evolution of deep-water mass composition at Hess Rise

The overall trend of ϵ_{Nd} values for site 464 shows a fundamental change in the composition of deep waters bathing Northern Hess over the time span of ~56 to ~32 Ma. From ~56 to ~51 Ma $\epsilon_{\text{Nd}}(t)$ values were relatively high, indicating the influence of waters from the North Pacific (Figure 3). The radiogenic values of the modern Pacific are due to weathering and drainage, from river water input, of arc volcanics in the circum-Pacific [Goldstein and Jacobsen, 1987]. These arc volcanoes were formed from the subduction of the Kula and Farallon plates that began ~110 Ma [Larson and Pitman, 1972]. Accordingly, it is likely that the North Pacific was the source for radiogenic deep-water Nd isotopic composition during the late Paleocene to early Eocene. This radiogenic signature of Nd sourced to the deep waters of the North Pacific is likely attributed to North Pacific deep-water production.

The Site 464 data indicate a shift to less radiogenic values (-3.02 to -4.38) from ~51 to ~32.3 Ma. The overall 4 epsilon unit decrease to less radiogenic values over the study interval suggests a relatively steady decrease in the influence of waters from the North Pacific. The lower, less radiogenic signature reflects a proportional increase in waters from the Southern Ocean [Thomas *et al.*, 2003; Thomas, 2004], although the location of Southern Ocean convection is still not clear. Despite a lack of understanding of Southern Ocean sourcing location, the Southern Ocean is the most likely source of the less radiogenic waters [Thomas, 2004].

The $\epsilon_{\text{Nd}}(t)$ values for site 464, follow the same general trend as the Shatsky Rise sites 1209 and 1211 recorded by Thomas (2004) (Figure 3), however there are noticeable

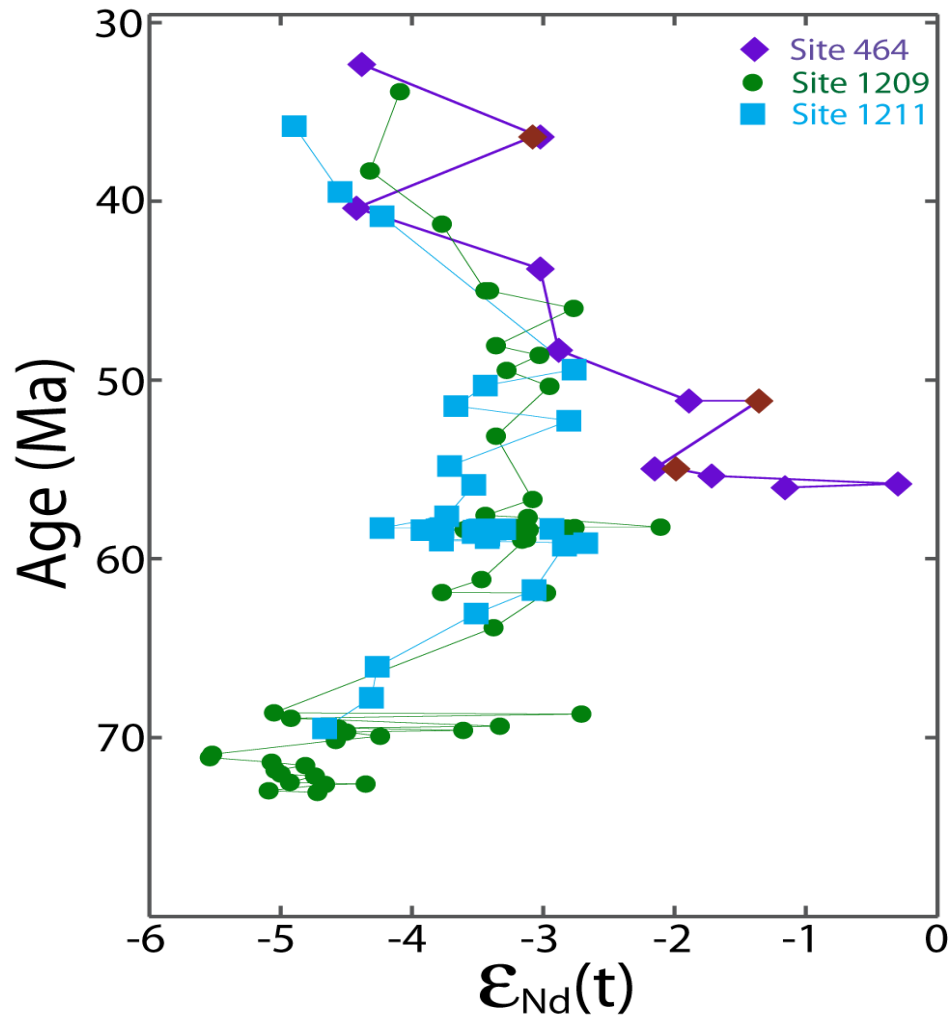


Figure 3. Site 464 data plotted with records from Sites 1209 and 1211 (Thomas, 2004). Cleaned samples colored red.

short term oscillations of varying magnitude in each site which is most likely attributed to sampling resolution. There are two interesting differences between the records. The first is that while the records from Sites 1209 and 1211 indicate a plateau in the composition for ~20 million years, the Site 464 data indicate a general decrease in the Nd isotope values. The temporal coverage of the Site 464 record does not completely overlap with Sites 1209 and 1211, but the available data suggest a somewhat different history of the water mass composition from 55 to 48Ma. After 48Ma all three sites record the same values and trends suggesting a common water mass source.

An important difference between Shatsky Rise and the northern Hess Rise lies in the strength of the radiogenic signature between Sites 1209/1211 and Site 464. Shatsky Rise data have a maximum $\epsilon_{Nd}(t)$ value of $-2.85 \sim 52.3$ Ma while site 464 has a maximum value of $-.30 \sim 55.8$ Ma. It is surprising to see such a difference in epsilon units between the two sites because they were located at the same latitude (paleolatitudes $\sim 18^\circ N$ for 464; $\sim 15^\circ N$ for 1211). Generally locations at the same latitude record similar epsilon values.

The large epsilon value difference (~ 2.0) between Shatsky Rise and Northern Hess Rise could imply that a separate water mass convected in the North Pacific from ~ 51 to 56 Ma. Site 464 is characterized by a paleodepth of 4600 m while sites 1209 and 1211 are characterized by paleodepths of ~ 2300 and ~ 2900 m respectively. There may have been a very radiogenic deep water mass that convected to depths of at least 4600 m but did not influence shallower depths, like that of sites 1209 and 1211, as significantly. This would imply that the North Pacific water mass was denser than the waters inferred

to have flowed northward from the Southern Ocean. Thus the shallower sites, although clearly influenced by the radiogenic water mass from ~65 to ~45Ma, did not record waters as radiogenic as the deeper site.

One interpretation of the data is that two water masses convected during this time, both sourced from the North Pacific Ocean based on the relatively radiogenic signatures recorded at all three sites. It is still unclear if there were in fact two water masses that convected from the North Pacific; hopefully future data will shed new light. Beginning ~49 Ma Shatsky and Northern Hess Rise record similar ϵ_{Nd} values indicating that a similar water mass began to bath sites 464, 1209 and 1211 (-2.9, -3.4, and -2.9), that most likely sourced from the Southern Ocean with a waning influence from the North Pacific source.

Another possible explanation for the difference of $\epsilon_{Nd}(t)$ values between Shatsky and Northern Hess Rise is the existence of the Emperor Seamount chain. Thus, the east-west geographic difference may have impacted the water mass composition at each location because Shatsky Rise lies to the west of the Emperor Seamount chain while Northern Hess Rise lies to the east. The seamounts in the chain were significantly shallower (some actually subaerially exposed) and buoyant ~55 Ma, and thus may have served as a bathymetric impediment to waters that may have convected in the northeastern Pacific. In this case, much of the deep-water radiogenic signature would not have reached Shatsky Rise, while Northern Hess Rise would have received more flow from the North Pacific deep waters. The trend back to more similar epsilon values between the sites beginning ~48.6 to ~32.3 Ma could be due to subsidence of the

Emperor Seamount over that time span, combined with diminishing convection in the North Pacific. As the chain subsided the deep-water signature recorded would be the same for both locations because the seamount no longer blocked the signature from North Pacific deep waters.

Leg 199 data from [Thomas *et al.*, 2008] show ϵ_{Nd} values between the ranges of -4 to -5. Compared to site 464 data these values are more radiogenic. However, Leg 199 sites were near the or below the paleoequator ~56 Ma, thus they were farther south than site 464. Such a difference in latitudinal location would expect a less radiogenic signature. The ϵ_{Nd} values remain relatively constant through time at sites 1215, 1217, 1219, and 1221 indicating that a common water mass bathed these sites and that the paleoequator was most likely the mixing location of Northern Pacific and Southern Ocean water masses [Thomas *et al.*, 2008]. The North Pacific water mass that convected to bath site 464 from ~56 to ~51 Ma was most likely the same water mass that mixed near the Leg 199 sites, indicating that as the water mass extended southward a less radiogenic signature was produced.

4.3. Implications of the new data

The future goal from the findings of new data from site 464 is to gain a better understanding on how the thermohaline circulation during the last greenhouse climate played a role in global heat transport. Model simulations will test circulation modes of deep water convection, as well as produce surface temperatures across the boundaries being studied. Then time slices coupled with ϵ_{Nd} values will be correlated with deep-water mass sourcing regions. The data generated in this study provides several

important constraints on the possible existence and influence of waters that convected in the North Pacific, and thus will be crucial to adapting upcoming model simulations.

4.4. Conclusions

The data from site 464 support the previous implications that there was a bimodal thermohaline circulation during the last major greenhouse climate of the early Cenozoic. The relatively more radiogenic signature (-.30 to -1.9) recorded from ~56 to ~51 Ma indicate that there possibly could have been two major water masses convecting during this time in the North Pacific; new data will hopefully lead to further justification of this assumption. It is also possible that the Emperor Seamounts could have affected the signature recorded between Shatsky and northern Hess Rise. Future studies will lead to continual understanding of the mode of the thermohaline circulation during the early Cenozoic, and shed new light on how this circulation drives global heat transport.

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